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[0004] A number of processes for the recovery or removal of carbon dioxide from gas streams have been proposed and practiced on a commercial scale. The processes vary widely, but generally involve some form of solvent absorption, adsorption on a porous adsorbent, distillation, or diffusion through a semipermeable membrane.

[0005] Membranes are thin barriers that allow preferential passage of certain components of a multi-component gas mixture. Most membranes can be separated into two types: porous and nonporous. Porous membranes separate gases based on molecular size and/or differential adsorption by small pores in the membrane. Gas separation membranes used in natural gas applications are often nonporous or asymmetric and separate gases based on solubility and diffusivity. These membranes typically have a microporous layer, one side of which is covered with a thin, nonporous "skin" or surface layer. The separation of the gas mixtures through an asymmetric membrane occurs in its skin, while the microporous substrate gives the membrane mechanical strength.

[0006] In a typical membrane separation process, a gas is introduced into the feed side of a module that is separated into two compartments by the permeable membrane. The gas stream flows along the surface of the membrane and the more permeable components of the gas pass through the membrane barrier at a higher rate than those components of lower permeability. After contacting the membrane, the depleted feed gas residue stream, the retentate, is removed from contact with the membrane by a suitable outlet on the feed compartment side of the module. The gas on the other side of the membrane, the permeate, is removed from contact with the membrane, the permeate side, through a separate outlet. The permeate stream from the membrane may be referred to as being "enriched" in the readily permeable components relative to the concentration of the readily permeable components in the retentate stream. The retentate may also be referred to as being "depleted" of the readily permeable components. While the permeate stream can represent the desired product, in most natural gas permeation processes the desired product is the retentate stream, and the permeate stream comprises contaminants such CO<sub>2</sub> or other acid gases.

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[0007] The efficiency of a membrane depends on many factors including the pressure differential being maintained across the membrane, whereby the permeable fluid component(s) permeate to the permeate side of the membrane under a partial pressure gradient. In order to maintain the partial pressure differential across the membrane, a sweep fluid is often used to help remove the permeating fluid. The lower the partial pressure of the permeate, the better the separation. This is especially important in applications where only small amounts of fluid are to be separated from the fluid mixture. However, many uses for the permeate require permeate further pressurization of the permeate. Low permeate partial pressure is desired for efficient membrane application, but high permeate pressure is desired to reduce compression costs.

[0008] While membrane systems that use sweep fluids have been effective in improving the efficiency of membrane separation of fluid, there is a continuing need for improving the efficiency of membrane separation processes.

### SUMMARY

[0009] This invention provides a method and system for separating at least one gaseous or vaporous component from a multi-component gas stream. A flow conduit is provided having a semi-permeable section adapted to selectively permeate the at least one gaseous component to be separated in the presence of the multi-component gas flowing along one side of the semi-permeable section. The multi-component gas is passed along the feed side of the flow conduit and a sweep gas, having a first velocity, is provided for passage along the permeate side of flow conduit, the sweep gas being suitable for sweeping the component gas that permeates through the permeable section of the conduit away from the permeate side of the flow conduit, thereby producing a gas mixture comprising the sweep gas and the component gas. The velocity of the sweep gas is accelerated so that the velocity of the sweep gas along at least a portion of the permeate side of the flow conduit is greater than the first velocity of the sweep gas. The gas mixture is then decelerated by means of a defuser, thereby recovering as pressure a portion of the energy of the gas mixture.

### **BRIEF DESCRIPTION OF THE DRAWINGS**

[0010] The present invention and its advantages will be better understood by referring to the following detailed description and the attached drawings, in which like reference numerals are used to indicate like parts in various views.

[0011] Fig. 1 is a sectional, schematic view of one embodiment of the present invention showing a feed gas conduit with a portion of the conduit's structure being a semi-permeable and a nozzle conduit surrounding a portion of the feed conduit for collecting permeate and for passing sweep gas across the semi-permeable structure at subsonic velocity.

[0012] Fig. 2 is a cross-sectional view of the embodiment shown in Fig. 1 taken along lines 2-2.

[0013] Fig. 3 is a sectional, schematic view of a second embodiment of the present invention similar to Fig. 1 except that the nozzle conduit surrounding the semi-permeable structure provides for supersonic velocity of the sweep gas across the semi-permeable structure.

[0014] Fig. 4 is a sectional, schematic view of a third embodiment of the present invention showing a nozzle conduit on the inside of a membrane conduit, the nozzle conduit collecting permeate and providing passage of sweep fluid at subsonic velocity.

### **DETAILED DESCRIPTION OF THE INVENTION**

[0015] The present invention provides an apparatus and method for separating one or more components from a multi-component gas using a separation system having a feed side and a permeate side separated by a semi-permeable structure. The separation system uses a sweep gas to facilitate removal of permeate from the permeate side of the separation system. This present invention increases the velocity

of sweep gas on the permeate side of the semi-permeable structure by reducing the cross-sectional area of sweep gas flow, thereby increasing the velocity of the sweep gas and reducing the static pressure of the permeate on the permeate side of the structure. The reduction in static pressure is achieved in one embodiment by using a converging nozzle for subsonic flow velocities and in another embodiment by using a converging-diverging nozzle for supersonic flow velocities.

[0016] The terms used in this description are defined as follows:

[0017] "Effuser" means a flow channel having a convergent section downstream of flowing section which functions as an aerodynamic expander. An effuser may have a converging volume or a converging and then diverging volume.

[0018] "Supersonic effuser" means a flow channel having a convergent subsonic section upstream of a divergent supersonic section with an intervening sonic throat which functions as an aerodynamic expander.

[0019] "Diffuser" means a flow channel having downstream divergent section which functions as an aerodynamic compressor. A diffuser have a diverging volume or a converging and then diverging volume.

[0020] "Supersonic diffuser" means a flow channel having a convergent super sonic section upstream of a divergent subsonic section with an intervening sonic throat which functions as an aerodynamic compressor.

[0021] "Throat" means a reduced area in a flow channel, as in an effuser or diffuser.

[0022] "Natural gas" refers to a multi-component gas obtained from a crude oil well (associated gas) or from a subterranean gas-bearing formation (non-associated gas). The composition and pressure of natural gas can vary significantly. A typical natural gas stream contains methane ( $C_1$ ) as a significant component. The natural gas will also typically contain ethane ( $C_2$ ), higher molecular weight hydrocarbons, one or more acid gases (such as carbon dioxide, hydrogen sulfide, carbonyl sulfide, carbon

[0034] The semi-permeable structure 25 for use in the present invention can be any suitable device having a selectively permeable nature, and more specifically it may be any device being relatively permeable to at least one component relative to one or more other components in the feed stream. The semi-permeable structure 25 can be of any suitable design for vapor separations. Tubular structures are preferred to obtain the benefits of the partial pressure reduction on the permeate side of the membrane in accordance with this invention. The semi-permeable structure 25 can be made entirely of the permselective material or the permselective material may be supported on a porous structure, fabric, or screen. The semi-permeable structure 25 is preferably composed of a separation layer and a support with the separation layer being formed on the surface of the support. The support is designed to provide mechanical support to the separation layer while offering as little mass transfer resistance as possible. The flux through the semi-permeable structure is affected by the thickness of the separation material and the support. In general it is desirable to have the separation layer, through which a permeating component must pass, as thin as possible yet sufficiently thick that the flow through the layer is not dominated by defects. The support must be thick enough to provide adequate strength to the separation layer to withstand the separation conditions. Suitable composite semi-permeable structures may comprise a thin separation layer or membrane formed on the surface of a thicker porous physical support that provides the necessary physical strength to the membrane. The number and length of the individual membranes used in the semi-permeable structure can be varied to suit the fluid flow rates and flux requirements of particular applications.

[0035] With respect to the composition of the separation layer, substantially any semi-permeable material currently available, or which may become available, can be used. The separation layer can be either symmetric or asymmetric, isotropic (having substantially the same density throughout) or anisotropic (having at least one zone of greater density than at least one other zone), and can be chemically homogenous (constructed of the same material) or it may be a composite membrane.

[0036] When the membrane separation systems illustrated Figs. 1 and 3 are used to remove contaminants from natural gas stream, the separation layer preferably is

composed of materials tolerant to temperatures above 120°F (48.9°C) and pressures above 1,200 psia (82.8 bar) and have adequate effective permeance and selectivity at those conditions. Many membranes in service for acid gas removal from natural gas streams are made from polymers, and most of these polymers either lack stability at the operating conditions at temperatures above 120°F (48.9°C) and pressures above about 1,200 psia (82.8 bar) or do not provide adequate values of permeance or selectivity. Many of such polymeric membranes have been designed or selected to operate most effectively at temperatures below about 100°F (37.8°C). While certain polymers or glassy materials could give adequate performance at higher temperature and pressure conditions, it is preferred that the separation layer used in natural gas treatment be inorganic. The inorganic layer, formed from, for example, zeolites, microporous silica, or microporous carbon, is preferably placed on a structured support.

[0037] The support should offer minimal mass transfer resistance with strength sufficient to withstand the stress created by relatively large pressure differentials across the membrane. For asymmetric membranes, the support is porous. It is also possible to form an asymmetric hybrid membrane structure in which a polymeric active separation layer is coated onto a porous inorganic support. For asymmetric inorganic membranes, the porous support can be made from a different material than the active separation layer. Support materials for asymmetric inorganic membranes include porous aluminas, silicon carbides, porous metals, cordierites, and carbons. Typically for asymmetric polymer membranes, the porous support is manufactured from the same polymer as the active separation layer. In some polymer membrane manufacturing processes, the porous support material is formed simultaneously with active separation layer.

[0038] The invention is not intended to be limited to any particular separation layer or support, and the separation layer and support may comprise any material capable of giving adequate values for permeance and selectivity. This includes, for example, homogeneous membranes, composite membranes, and membranes incorporating sorbents, carriers, or plasticizers. Inasmuch as the composition and

preparation of membrane are well known to those skilled in the art, a more detailed description thereof is not provided herein.

[0039] Figs. 1 and 3 illustrates embodiments in which the multi-component fluid to be treated and the sweep fluid are in countercurrent flow, which is the preferred arrangement. However, co-current arrangements could also be produced, one embodiment of which is shown in Fig. 4.

[0040] Fig. 4 illustrates a sectional, schematic view of a third embodiment of the present invention showing an effuser 40 and diffuser 41 on the inside of a multi-component feed gas stream that is passing through a flow conduit 42 in the direction of arrow 43. A semi-permeable membrane module 44 is disposed on the inside of a flow conduit 42. Membrane module 44 comprises a membrane layer 45 that is coated or bonded to the surface of a support member 46. The membrane module 44 may also include other layers not shown in Fig. 4, such as a protective layer that may include for example a cage or screen to protect the outside membrane layer.

[0041] Sweep gas 49 enters the separation module through sweep gas inlet conduit 48. The direction of sweep gas 49 into conduit 48. Inlet conduit 48 passes through the bulb-shaped end of membrane module 44 and ends at nozzle 50, thereby enabling sweep gas 49 to flow through inlet conduit 48 and exit through nozzle 50. The velocity of sweep gas 49 through nozzle 50 induces a low pressure zone in the throat 51 of a venturi portion 52 of the membrane module 44, drawing permeate through the membrane layer 45 to the interior of membrane module 44. A diffuser 41 is located downstream of the membrane module 44. The high velocity of the mixture of permeate and sweep gas exiting the membrane module 44 is reduced in velocity in diffuser 41 to produce an increase in pressure over that of the gas mixture through section venturi portion 52.

[0042] In Fig. 4, diffuser 41 is shown as being positioned immediately following the passage of the sweep gas past membrane module 44. However, the diffuser 41 may optionally be positioned farther downstream than shown in Fig. 4. The diffuser 41 may optionally be outside conduit 42.

[0043] The method of the present invention may be practiced in any flow environment involving two or more concentric flow lines in which at least a portion of

**Box No. VIII (iii) DECLARATION: ENTITLEMENT TO CLAIM PRIORITY\***

*The declaration must conform to the standardized wording provided for in Section 213; see Notes to Boxes Nos. VIII, VIII (i) to (v) (in general) and the specific Notes to Box No. VIII (iii). If this Box is not used, this sheet should not be included in the request.*

Declaration as to the applicant's entitlement, as at the international filing date, to claim the priority of the earlier application specified below, where the applicant is not the applicant who filed the earlier application or where the applicant's name has changed since the filing of the earlier application (Rules 4.17(iii) and 51bis.1(a)(iii)):

In relation to this International Application

KELLEY, Bruce T. is entitled to claim priority of earlier Application

No. 60/413,350 filed 25 September 2002 (25.09.02)

by virtue of the following:

the applicant is the Inventor of the subject matter from which protection was sought by way of the earlier application.

This declaration is made for the purposes of the following designations for national and/or regional patents: US.

☐ This declaration is continued on the following sheet, "Continuation of Box No. VIII (iii)".